This Page Is Inserted by IFW Operations and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

As rescanning documents will not correct images, please do not report the images to the Image Problem Mailbox.

THIS PAGE BLANK (USPTO)

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51)	International Patent Classification: C08G 18/00	A2	1 ()	tional Publication Number: tional Publication Date:	WO 00/73362 07 December2000 (07.12.2000)
(21)	International Application Number:	PCT/	EP00/04038	Published	
(22)	International Filing Date: 05 May	2000	(05.05.2000)	rubiisileu	
(30)	Priority Data: 99110480.3 31 May 1999 (31.05	.1999)) EP		
(60)	Parent Application or Grant HUNTSMAN ICI CHEMICALS, LLC [/]; Alain [/]; (). YU, Jianming [/]; (). PARFON (). YU, Jianming [/]; (). BAKEN, Philippus Leonardus, Henricus @; ().	√ĎRY	, Alain [/];		

- (54) Title: PROCESS FOR MAKING LOW DENSITY FOAMS, POLYOL COMPOSITION AND REACTION SYSTEM USEFUL THEREFOR
- (54) Titre: PROCESSUS DE FABRICATION DE MOUSSES DE FAIBLE DENSITE, COMPOSITION AU POLYOL ET SYSTEME DE REACTION A CET EFFET

(57) Abstract

The invention relates to a polyol composition comprising by weight 60-97 % of b1) a polyoxyethylene-polyoxypropylene polyol, having a functionality of 2-6 with 10-25 % tipped EO; 3-40 % of b2) a polyoxyethylene-polyoxypropylene polyol, having a functionality of 2-6, with 20-50 % total EO and 10-20 % tipped EO; and 0-25 % of b3) a polyol, having a functionality of 2-6, with at least 50 % random EO. The invention also relates to a process for preparing a flexible polyurethane foam by reacting a) a polyisocyanate composition; b) a polyol composition of the invention; c) water, and d) additives and auxiliaries known per se. The invention finally relates to a reaction system comprising A) a polyisocyanate and B) an isocyanate-reactive component comprising the polyol of the invention.

(57) Abrégé

Cette invention concerne une composition au polyol, qui comprend en masse 60 à 97 % b1) d'un polyoxyéthylène-polyoxypropylène polyol, ayant une fonctionnalité de 2 à 6 avec 10 à 25 % d'EO d'extrémité de chaîne; 3 à 40 % en masse b2) d'un polyoxyéthylène-polyoxypropylène polyol, ayant une fonctionnalité de 2 à 6, avec 20 à 50 % d'EO total et 10 à 20 % d'EO d'extrémité de chaîne; 0 à 25 % en masse b3) d'un polyol, ayant une fonctionnalité de 2 à 6, avec au moins 50 % d'EO aléatoire. Cette invention concerne aussi un processus de confection d'une mousse de polyuréthanne souple que l'on obtient en faisant réagir a) une composition polyisocyanate; b) une composition au polyol de l'invention; c) de l'eau; et d) des additifs et des agents auxiliaires connus per se. Cette invention concerne enfin un système de réaction qui comprend A) un polyisocyanate et B) un composant réagissant aux isocyanates comprenant le polyol de l'invention.

(19) World Intellectual Property Organization International Bureau



(43) International Publication Date 7 December 2000 (07.12.2000)

PCT

(10) International Publication Number WO 00/73362 A2

(51) International Patent Classification7:

C08G 18/00

- (21) International Application Number: PCT/EP00/04038
- (22) International Filing Date: 5 May 2000 (05.05.2000)
- (25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data: 99110480.3

31 May 1999 (31.05.1999) EP

(71) Applicant (for all designated States except US): HUNTS-MAN ICI CHEMICALS, LLC [US/US]; 500 Huntsman Way, Salt Lake City, UT 84108 (US).

(72) Inventors; and

- (75) Inventors/Applicants (for US only): PARFONDRY, Alain [FR/BE]; Rue Longue 82, B-1150 Bruxelles (BE). YU, Jianming [CN/BE]; Avenue Charles Woeste 66, B-1090 Bruxelles (BE).
- (74) Agents: BAKEN, Philippus, Johannes, Leonardus, Henricus et al.; Huntsman ICI Europe Ltd, Huntsman Polyurethanes, Intellectual Property Dept., Everslaan 45, B-3075 Everberg (BE).

- (81) Designated States (national): AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, IP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TI, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

 Without international search report and to be republished upon receipt of that report.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

42

(54) Title: PROCESS FOR MAKING LOW DENSITY FOAMS, POLYOL COMPOSITION AND REACTION SYSTEM USE-FUL THEREFOR

(57) Abstract: The invention relates to a polyol composition comprising by weight 60-97 % of b1) a polyoxyethylene-polyoxypropylene polyol, having a functionality of 2-6 with 10-25 % tipped EO; 3-40 % of b2) a polyoxyethylene-polyoxypropylene polyol, having a functionality of 2-6, with 20-50 % total EO and 10-20 % tipped EO; and 0-25 % of b3) a polyol, having a functionality of 2-6, with at least 50 % random EO. The invention also relates to a process for preparing a flexible polyurethane foam by reacting a) a polyisocyanate composition; b) a polyol composition of the invention; c) water; and d) additives and auxiliaries known per se. The invention finally relates to a reaction system comprising A) a polyisocyanate and B) an isocyanate-reactive component comprising the polyol of the invention.

Description

15 -

PROCESS FOR MAKING LOW DENSITY FOAMS, POLYOL COMPOSITION AND REACTION SYSTEM USEFUL THEREFOR

The instant invention relates to a process for making low density foams. It also relates to a specific polyol composition and to a reaction system that are useful in the said process.

For flexible polyurethane foams, low density means cost effectiveness. Thus, low density is a target, that should however not be obtained with detrimental effects on the foam properties. Physical blowing, using carbon dioxide for example, is known to reduce foam density, but is associated with processing difficulties as well as the need for additional equipment.

US-P-5 686 502 discloses foams obtained through a one-shot process, where the polyol comprises a first polyol which is a poly(oxyalkylene)triol which is chain terminated with oxypropylene (PO) and a second polyol which is either (i) a poly(oxyalkylene)diol terminated with EO, or (ii) a polyfunctional polyol terminated with PO. The thus obtained foams are hydrophylic. The densities obtained in the examples vary between 13 and 20 kg/m³. There is no mention of the resilience.

Which are prepared by reacting a specific polyol composition. The polyol composition comprises a block PO/EO polyol having an OH value of 14 to 65, 2 to 9 % of tipped EO and a functionality of 2.3 to 2.8 and a di- or tri-functional PO/EO polyol having an OH value of 20 to 80 and 60 to 85 % of EO (preferably up to 20 % as tipped EO). The resulting foams are visco-elastic and do not exhibit any ball rebound (for densities obtained in the examples of about $70-77~{\rm kg/m^3}$).

US-P-4 833 176 discloses a process comprising reacting a polyisocyanate with a polyol at a NCO index below 70. The polyol may vary; examples comprise mixtures of a low EO-content polyol and high EO content polyol.

PCT/EP00/04038 WO 00/73362 2

5

10

15

20

25

30

35

40

45

50

55

10

15

20

25

30

EP-A-0 845 485 discloses a process for preparing flexible foams comprising reacting a polyisocyanate with a polyol, where the polyol is a specific polyol composition. Said polyol composition comprises: (i) a polyetherpolyol having a functionality of 2.5-6.0, which is a PO/EO polyol with 15% or less of EO; (ii) a polyetherpolyol having a functionality of 1.8-2.5, which is an all-PO polyol; (iii) a polyetherpolyol having a functionality of 1.8-6.0 and having an EO content of at least 50 wt%. The respective amounts of components (i), (ii) and (iii) are as follows: (i) 15-70 %, (ii) 30-80 %, and (iii) 3-15 %, based on the combined weights of the polyols.

US-P-5 594 097 discloses a polyol comprising PO and EO, having an OH value of 16-45, a primary hydroxyl content of at least 50 %, an EO content of 21-49 %, and having a structure of the type PO-(PO/EO)-EO, where the tipped EO content is 10-20 %. This specific polyol is said to be usable in combination with other polyols. All however, relate to polyol compositions examples, comprised solely of this specific polyol. Also, while a relatively low density is obtained, (i) there is no disclosure of the resilience values and (ii) there are processing difficulties and bad compression set when the EO-enriched polyol is used as the main polyol.

None of the above documents teaches or suggests the instant invention.

The following way of describing polyols is used in the present application : A PO-EO polyol is a polyol having first a PO block attached to the initiator followed by an EO block. A PO-PO/EO polyol is a polyol having first a PO block and then a block of randomly distributed PO and EO. A PO-PO/EO-EO polyol is a polyol having first a PO block then a block of randomly distributed PO and EO and then a block of EO. polyol is a polyol having first a PO block and then an EO In the above descriptions only one tail of a block.

polyol is described (seen from the initiator); the 5 nominal hydroxy functionality will determine how many of such tails will be present. The present invention provides a process that surprisingly affords a resilient flexible polyurethane 10 foam having a good stability (low recession) and the advantages of EO enriched polyol as the main polyol (i.e. density reduction) without having the drawbacks (i.e. negative impact on mechanical properties, like tensile 15 strength, elongation and tear strength). 10 The invention thus provides a polyol composition comprising : polyoxyethylene-polyoxypropylene polyol, b1) а 20 having an average nominal hydroxyl functionality of 2-6 where the EO is present as tipped EO, the EO content 15 being between 10-25 % by weight based on the weight of the polyol 25 polyoxyethylene-polyoxypropylene polyol, b2) a having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and random EO, the 20 total EO content being between 20-50 % and the tipped EO content being between 10-20 %, both by weight based on 30 the weight of the polyol a polyol, having an average nominal hydroxy b3) functionality of 2-6, and comprising EO and optionally PO 25 where the EO is present as random EO, the EO content 35 being at least 50 % by weight based on the weight of the polyol these polyols bl, b2 and b3 being present according to the following proportions, based on the combined 40 30 weights of b1, b2 and b3, b1 : 60-97 wt %, b2 : 3-40 wt %, b3 : 0-25 wt %. Unless otherwise stated amounts of EO and PO in a polyol are indicated hereinafter as % by weight based on 45 the weight of the polyol. 35

PCT/EP00/04038

4

5 The invention thus provides a process for preparing a flexible polyurethane foam at an NCO index of 70-120 and preferably of 70-105 by reacting: a) a polyisocyanate; 10 5 polyoxyethylene-polyoxypropylene having an average nominal hydroxyl functionality of 2-6 where the EO is present as tipped EO, the EO content being between 10-25 %; polyoxyethylene-polyoxypropylene 15 having an average nominal hydroxy functionality of 2-6, 10 where the EO is present as tipped EO and random EO, the

WO 00/73362

20

25

30

35

40

45

50

55

15

20

25

30

35

where the EO is present as tipped EO and random EO, the total EO content being between 20-50 %, the tipped EO content being between 10-20 %,

b3) a polyol, having an average nominal hydroxy

functionality of 2-6, and comprising EO and optionally PO where the EO is present as random EO, the EO content being at least 50 %,

these polyols b1, b2 and b3 being present according to the following proportions, based on the combined weights of b1, b2 and b3, b1 : 60-97 wt %, b2 : 3-40 wt %, b3 : 0-25 wt %;

c) water; and

d) additives and auxiliaries known per se.

The invention finally relates to a reaction system comprising A) a polyisocyanate and B) an isocyanate-reactive component comprising the polyol of the invention and water, as well as to a reaction system comprising A) a polyisocyanate prepolymer obtained by reacting the polyisocyanate with part of the polyol composition of the invention, and B) an isocyanate-reactive component comprising the remainder of the polyol composition of the invention and water.

In the context of the present invention the following terms, if and whenever they are used, have the following meaning:

1) isocyanate index or NCO index :

the ratio of NCO-groups over isocyanate-reactive 5 hydrogen atoms present in a formulation, given as a percentage : [NCO] \times 100 (8) 10 5 [active hydrogen] In other words the NCO-index expresses the percentage of isocyanate actually used in a formulation with respect to the amount of isocyanate theoretically required for 15 reacting with the amount of isocyanate-reactive hydrogen 10 used in a formulation. It should be observed that the isocyanate index as used herein is considered from the point of view of the 20 involving isocyanate the process foaming 15

It should be observed that the isocyanate index as used herein is considered from the point of view of the actual foaming process involving the isocyanate ingredient and the isocyanate-reactive ingredients. Any isocyanate groups consumed in a preliminary step to produce modified polyisocyanates (including such isocyanate-derivatives referred to in the art as quasi or semi-prepolymers and prepolymers) or any active hydrogens reacted with isocyanate to produce modified polyols or polyamines, are not taken into account in the calculation of the isocyanate index. Only the free isocyanate groups and the free isocyanate-reactive hydrogens (including those of the water, if used) present at the actual foaming stage are taken into account.

2) The expression "isocyanate-reactive hydrogen atoms" as used herein for the purpose of calculating the isocyanate index refers to the total of hydroxyl and amine hydrogen atoms present in the reactive compositions in the form of polyols, polyamines and/or water; this means that for the purpose of calculating the isocyanate index at the actual foaming process one hydroxyl group is considered to comprise one reactive hydrogen, one primary or secondary amine group is considered to comprise one reactive hydrogen and one water molecule is considered to comprise two active hydrogens.

50

25

30

35

40

45

20

25

30

PCT/EP00/04038 WO 00/73362 6

5		3) Reaction system : a combination of components
		wherein the polyisocyanate component is kept in a
		container separate from the isocyanate-reactive
!		components.
10	5	4) The expression "polyurethane foam" as used
		herein generally refers to cellular products as obtained
		by reacting polyisocyanates with isocyanate-reactive
		hydrogen containing compounds, using foaming agents, and
15		in particular includes cellular products obtained with
	10	water as reactive foaming agent (involving a reaction of
		water with isocyanate groups yielding urea linkages and
		carbon dioxide and producing polyurea-urethane foams).
20		 The term "average nominal hydroxyl
		functionality" is used herein to indicate the average
	15	functionality (number of hydroxyl groups per molecule) of
	-	the polyol composition on the assumption that this is the
25		average functionality (number of active hydrogen atoms
		per molecule) of the initiator(s) used in their
		preparation although in practice it will often be
	20	somewhat less because of some terminal unsaturation.
30		6) The term "average" is used to indicate an
		average by number.
		The polyisocyanates may be selected from aliphatic,
		cycloaliphatic and araliphatic polyisocyanates,
35	25	especially diisocyanates, like hexamethylene
-		diisocyanate, isophorone diisocyanate, cyclohexane-1,4-
		diisocyanate, 4,4'-dicyclohexylmethane diisocyanate and
		m- and p-tetramethylxylylene diisocyanate, and in
40		particular aromatic polyisocyanates like tolylene
	30	diisocyanates (TDI), phenylene diisocyanates and most
		preferably diphenylmethane diisocyanate optionally
		comprising homologues thereof having an isocyanate
45		functionality of 3 or more (such diisocyanates comprising
70		such homologues are known as crude MDI or polymeric MDI
	35	or mixtures of such crude or polymeric MDI with MDI) and
		modified variants thereof.

50

7

The diphenylmethane diisocyanate (MDI) used may be selected from 4,4'-MDI, 2,4'-MDI, isomeric mixtures of 4,4'-MDI and 2,4'-MDI and less than 10% by weight of and modified variants thereof containing 2,2'-MDI, urethane, isocyanurate, uretonimine, carbodiimide, allophanate, urea and/or biuret groups. Preferred are 4,4'-MDI, isomeric mixtures of 4,4'-MDI and 2,4'-MDI and less than 10% by weight of 2,2'MDI and uretonimine and/or carbodiimide modified MDI having an NCO content of at least 20% by weight and preferably at least 25% by weight and urethane modified MDI obtained by reacting excess MDI and polyol having a molecular weight of at most 1000 and having an NCO content of at least 20% by weight and preferably at least 25% by weight.

15

25

30

35

10

5

10

15

20

25

30

35

40

Diphenylmethane diisocyanate comprising homologues having an isoycanate functionality of 3 or more are so-called polymeric or crude MDI.

Polymeric or crude MDI are well known in the art.

They are made by the phosgenation of a mixture of polyamines obtained by the acid condensation of aniline and formaldehyde.

The manufacture of both the polyamine mixtures and the polyisocyanate mixtures is well known. condensation of aniline with formaldehyde in the presence of strong acids such as hydrochloric acid gives a diaminodiphenylmethane containing product together with polymethylene polyphenylene polyamines of higher functionality, the precise composition depending in known manner inter alia on the aniline/formaldehyde The polyisocyanates are made by phosgenation of the polyamine mixtures and the various proportions of diamines, triamines and higher polyamines give rise to related proportions of diisocyanates, triisocyanates and The relative proportions of higher polyisocyanates. diisocyanate, triisocyanate and higher polyisoycanates in

50

5

10

15

20

25

30

35

40

45

50

55

25

30

such crude or polymeric MDI compositions determine the average functionality of the compositions, that is the average number of isocyanate groups per molecule. varying the proportions of starting materials, average functionality of the polysiocyanate compositions can be varied from little more than 2 to 3 or even In practice, however, the average isocyanate functionality preferably ranges from 2.3-2.8. value of such polymeric or crude MDI is at least 30% by 10 MDI weight. The polymeric or crude contain diisocyanate, the remainder being diphenylmethane polyisocyanates polymethylene polyphenylene functionality greater than two together with by-products formed in the manufacture of such polyisocyanates by phosgenation of polyamines. Further modified variants of 15 such crude or polymeric MDI may be used as well uretonimine, carbodiimide, isocyanurate, comprising urea and/or biuret groups; urethane, allophanate, uretonimine especially the aforementioned carbodiimide modified ones and the urethane modified ones 20 are preferred. Mixtures of polyisocyanates may be used as well.

The invention also relates to a polyol composition, comprised of polyols b1, b2 and b3.

Polyol bl can be prepared by known methods. It has a structure of the type PO-EO, where EO is present as tipped EO. The EO content is from 10 to 25 % by weight.

Polyol b2 can also be prepared by known methods. It can have a structure of the type PO-PO/EO-EO or of the type PO/EO-EO. EO is present as tipped and random. The total EO content is from 20 to 50 % by weight, preferably from 21 to 49 %, the tipped EO content is from 10-20 % by weight. In the PO-PO/EO-EO type polyol, the first PO block comprises preferably from 20 to 75 % by weight of the PO units. Preferably the weight ratio tipped EO/random EO is from 1:3 to 3:1. The polyol having a

structure of the type PO-PO/EO-EO can notably be produced 5 according to the teaching of US 5594097. The polyol having a structure of the type -PO/EO-EO can notably be produced according to the teaching of US 4559366. Polyol b3 is the optional polyol. It can also be 10 5 prepared by known methods. It can have a structure of type PO/EO or of the type -EO (PEG). EO is present as random EO (if and when PO is present). The EO content is Preferably it more than 50% by weight. 15 The functionality of these polyoxyethylene polyol. 10 polyols is comprised between 2 and 6, preferably between 2 and 4. For bl and b2, the equivalent weight is generally 20 comprised between 1000 and 4000, preferably 1500 and 3500; while for b3, the equivalent weight is generally 15 comprised between 200 and 3000, preferably 300 and 2000. The polyol composition comprises the various polyols 25 according to the following proportions, expressed on the basis of the combined weights of the polyols: b1 : 60-97 %, preferably 65-90 % 20 3-40 %, preferably 10-30 % b2: 30 0-25 %, preferably 0-10 % (more preferably 3-10 %); all percentages being % by weight. Each component b1, b2 and b3 may be comprised of mixtures. 25 Dispersed material can also be present. This is known 35 as polymer-modified polyol, and comprise e.g. SAN or PIPA (Poly Isocyanate Poly Addition). The polymer-modified polyols which are particularly interesting in accordance with the invention are products 40 30 obtained by in situ polymerisation of styrene and/or poly(oxyethylene/oxypropylene)polyols acrylonitrile in and products obtained by in situ reaction between a polyisocyanate and an amino- or hydroxy-functional 45 triethanolamine) compound (such as 35 poly(oxyethylene/oxypropylene)polyol. The solids content (based on the total polyol weight b1+b2+b3) can vary

PCT/EP00/04038 WO 00/73362 10

5

10

15

20

25

30

35

40

45

50

55

10

15

20

25

30

35

within broad limits, e.g. from 5 to 50 % by weight. Particle sizes of the dispersed polymer of less than 50 microns are preferred. Mixtures can be used as well.

Water is used as the blowing agent. Carbon dioxide may be added if needed. In the case of highly resilient water blown flexible foams, it is appropriate to use from 1.0 to 15 and preferably from 2 to 10 % by weight of water based on the weight of the total polyol component where the water can optionally be used in conjunction with carbon dioxide.

Other conventional ingredients (additives and/or auxiliaries) may be used in making the polyurethanes. These include catalysts, for example, tertiary amines and organic tin compounds, surfactants, cross linking or chain extending agents, for example, low molecular weight compounds such as diols, triols (having a molecular weight below the one of b3) and diamines, flame proofing agents, for example halogenated alkyl phosphates, fillers and pigments. Foam stabilizers, for example polysiloxanepolyalkylene oxide block copolymers, may be used to stabilize or regulate the cells of the foam.

The amount of these minor ingredients used will depend on the nature of the product required and may be varied within limits well known to a polyurethane foam technologist.

The present invention also relates to a process for preparing a flexible polyurethane foam at an NCO index of polyisocyanate a); 70-120 by reacting а polyoxyethylene-polyoxypropylene polyol b1); polyoxyethylene-polyoxypropylene polyol b2); a polyol b3); according to ratios specified above; water c); and additives and auxiliaries known per se d).

These components, notably the polyols b1, b2 and b3 can be added in any order. Notably, the polyols can be added according to the following non-limiting possibilities:

Part of b1+b2+b3, then the remainder of b1+b2+b3;

5		Part of b1+b2 but no b3, then the remainder of
		b1+b2 and all b3;
		Part of b1+b3 but no b2, then the remainder of
		b1+b3 and all b2;
10	5	all of b1, then the all of b2+b3; all of b2, then
		the all of b1+b3;
		Part of b1, then the remainder of b1 together
		with the all of b2+b3;
15		Part of b2, then the remainder of b2 together
	10	with the all of b1+b3;
		And any other possibility.
		In the process of the invention, it is to be noted
20		that one shot, prepolymer or quasi-prepolymer methods may
		thus be employed as may be appropriate for the particular
,	15	type of polyurethane being made. The components of the
		polyurethane forming reaction mixture may be mixed
25		together in any convenient manner, for example the
20		individual components may be pre-blended so as to reduce
		the number of component streams to be brought together in
	20	the final mixing step. It is often convenient to have a
30		two-stream system whereby one stream comprises a
30		polyisocyanate or isocyanate-terminated prepolymer and
		the second stream comprises all the other components of
		the reaction mixture.
35	25	The flexible foams may be made according to
33		techniques known in the art like the moulding or the
		slabstock technique. The loams may be accoming
		furniture and automotive industry
40		cushioning and mattresses.
40	30	The flexible foams thus obtained have a free rise
		density comprised between 18 and 60 kg/m ³ . These foams
		show a resilience higher than 45 %. The following examples illustrate the invention
45		The following examples illustrate
45		without limiting same. Unless otherwise indicated, all parts are given by
	35	
		weight.

5	Glossary	
		onalities are nominal functionalities,
	_	ights are nominal equivalent weights, all %
	are % by weigh	nt and OH values are in mg KOH/g)
10	Polyol A	PO-EO, with EO as tipped. EO content is
		15 %. Equivalent weight is 2004.
		Functionality is 3, OH value is 28.
	Polyol B	PO-PO/EO-EO, total EO content is 21 %. Tip
15		EO content is 15 %. Equivalent weight is
		2004. Functionality is 3, OH value is 28.
	Polyol C	PO-PO/EO-EO, total EO content is 28,6 %.
		Tip EO content is 15 %. Equivalent weight
20		is 2004. Functionality is 3, OH value is
		28.
	Polyol D	PO/EO-EO, total EO content is 26 %. Tip EO
		content is 15 %. Equivalent weight is 2158.
25		Functionality is 3, OH value is 26.
	Polyol E	PO/EO-EO, total EO content is 21 %. Tip EO
		content is 15 %. Equivalent weight is 1934.
		Functionality is 3, OH value is 29.
30	Polyol F	polyoxyethylene polyol having an equivalent
		weight of 450, an OH value of 123 and a
		functionality of 3.
	Polyol G	Polymer polyol, comprising 25 % of
35		dispersed particulate SAN material in high
33		molecular weight polyol, similar to polyol
		A, but with an equivalent weight of 1600
		and an OH value of 35.
40	Polyol H	PO/EO-EO, total EO content is 28 %. Tip EO
40		content is 15 %. Equivalent weight is 2004.
		Functionality is 3, OH value is 28. Primary
		hydroxyl content is 85.2
	Polyol I	PO-PO/EO-EO, total EO content is 28 %.
45		First PO block contains 55% PO over total
		PO and EO. Tip EO content is 15 %.
		Equivalent weight is 2004. Functionality is
		3, OH value is 29. Primary hydroxyl content
50		

_			
5		_	is 86.7 MDI comprising 93.8 % diisocyanate 48.2 %
		Isocyanate A	of which is 2,4'-MDI and 6.2 % is oligomer
			species of higher functionality.
10	•		Functionality is 2.05.
		Isocyanate B	MDI comprising 87.5 % disocyanate 46.0 %
			of which is 2,4'-MDI and 12.5 % is oligomer
			species of higher functionality.
15			Functionality is 2.10.
		Isocyanate C	Quasi-prepolymer based on MDI (81.3 %
		•	diisocyanate 30 % of which is 2,4'-MDI and
			18.7 % is oligomer species of higher
20			functionality, Functionality is 2.16.) and
			polyol A. NCO value is 29.7.
		Isocyanate D	MDI comprising 78.2 % diisocyanate 26.0 %
			of which is 2,4'-MDI and 21.8 % is oligomer
25			species of higher functionality.
	-		Functionality is 2.19.
		D8154	Amine catalyst from Air Products
		Niax Al	Catalyst from Union Carbide
30		D33LV	Catalyst from Air Products
30		DMEA	Dimethylethanolamine
		DETDA	diethyl toluenediamine
		Foams are	e produced according to the following scheme.
0.5		Polyols, cata	alysts, surfactants, water are mixed prior to
35		the addition	of isocyanates. Polyol blends and isoyanates
		are mixed at	20°C during 8 seconds before foaming. Free-
	, 5	rise foams a	re made in plastic buckets of 2.5 l to 10 l.
		Moulded foar	ns are made with a square mould of 9.1 l
40		preheated to	45°C.
			perties of the foam are determined according
		to the follo	wing methods and standards :
	10	FRD (Free Ri	se Density);
45		OAD (OverAll	Density) (kg/cm³), and CD (Core Density)
		(kq/cm^3) :	ISO 845
		Compression	hardness; CLD 40% (kPa) and Hysteresis Loss
		(%) :	ISO 3386-1
50			

PCT/EP00/04038 WO 00/73362

14 5 Compression set (thickness) : Dry 75 % (%) and Humid 75 % (%): Indentation Hardness :ILD 40 % (N) and Hysteresis Loss 10 (%): ISO 2439 5 Resilience (%) Toyota ISC 8067 Tear strength, max (N/m): Tensile strength(kPa) and Elongation (%): ISO 1798 15 The results are summarized in the following tables. 10 From the last table, one will note that the specific polyols of the type PO/EO-EO are even better than those of the type PO-PO/EO-EO, since they provide higher foam 20 stability (lower recession %) and lower free rise 15 density, and are thus particularly designed for making lower density foams. 25 30

35

40

45

50

						Examples	ples					
	-	,	~	4	2	9	7	80	6	10	11	12
Component	4	7	, ,	L	9.5	25	65	60	85	7.5	75	65
Polyol A	7.5	69	60	6/	3	5	3					
Polyol B	20	30	30	20				,	,		ç	200
Polvol C					10	10	30	35	OT I	02	77	3
Polvol D												
Polvol E										_	u	5
Polyol F	5	5	2	2	2	2	2	n		,	,	,
Polvol G									-		1	α
Water	6.5	6.5	9	9	2		9	٥	2	٥	,	3
4113	Œ C	8	1.2	1.2	0.5	1.0	1.0	1.0	1.0	1.0	1.0	0
113	, ,	7 0	9	9.0	0.7	0.5	0.5	0.5	0.5	0.5	0.5	0.5
8154)					0	0.1	0.1	0.1	0.1	0.1	0.1
Niax Al	0.1	U.1	1.0	,			2	6	7	7	٠	0.3
33 LV	0.3	0.3	0.3	0.3		ر اد	0.3	2		3	2	
OMEA					0.5							
ETDA					0.3			١				
Socvenate A	80	80				75	85	8.5			0	0
Tsocyanate B			80	80					75	85	82	001
Teoryanate C					97				,	,		0
Sold in College	Ca	Oα	98	98	85	83	85.6	85.6	96	86.4	BU BU	2.78

5					_						_	1	6					_,	- 7	_
·		24	55				20	5	20	4	0.5	0.7			0.5	0.3			63	86
. 10		23	55			20		5	20	4	0.5	0.7			0.5	0.3			63	86
		22	65			1.0		5	20	4	0.5	0.7			0.5	0.3			63	98
15		21	55	30				2	15	9.9	1.2	9.0	0.1	0.3				85		83.5
20		20	29		8			2	20	. 2	0.5	0.7			0.5	0.3			76	86
	les	19	67		8			5	20	4	0.5	0.7			0.5	0.3			63	98
25	Examples	18	85		10			5		ı,	1.2	9.0	0.1	0.3				08		101
30		17	85		10			5		5	1.2	9.0	0.1	0.3				75		96
		16	65	30				5		7	1.2	9.0	0.1	0.3				85		79
35		15	65	30				5		7	1.2	9.0	0.1	0.3				08		74
40		14	65	30				5		9	1.2	9.0	0.1	0.3				80		98
		13	65	30				2		و	1.2	9.0	0.1	0.3				75		80
45		nent	1 A	1 B		1	1	1	1 6	1	E7	14	A1	ΓΛ			Isocyanate A	Isocvanate B		NCO index
50		Component	Polyol	Polyol	Polvol	Polvol	Polvol	Polyo	Polvol	Water	B 4113	D 8154	Niax Al	D 33 LV	DMEA	DETDA	Isoc	Isoc	Isoc	NCO

•	.,	٠	

				_								17					_	_				,	
	12	oben	7	19.6					3.2	46.5													
	11	oben	.0	21.2	1			T	2.2	42.4							48.2						
	10	open*	.0	19.6	,				6	. o.	2												
	6	*nago	2 0 0 0 2	, r,	23:3				1 7	2.8 2.0 2 2.7	77.						55 1	5					
	8	open	,	1 6	23.3				,	12.7	17.												
les	7	neuo	,	7 60 7 70 11 10	0:47				o	20.0	37.3												
Examples	9	nacco		, ,	/./>				c	۵۰۶	24												
	5	0000	200	۲ ر	33.5				ר	3.23	35.05												
	4	* "0"	oben oben open open))	22.8																		
	~	4	obeil	э ;	22				ţ	1.7	34.3												
	6		obeu	7	23.3																		
	-	1	oben	∞	26.7																		
		Properties	Cells	Recession %	FRD (kg/cm ³)	Moulding	overall density kg/m³)	Core density (kg/m³)	Compression hardness	CLD 40 % (kPa)	Hysteresis (%)	Compression set (thick)	Dry 75 % (%)	Humid 75 % (%)	Indentation hardness	ILD 40 % (N)	Hysteresis (%)	Resilience (%)	Tear strength	Max (N/m)	Tensile strength (kPa)	Elongation (%)	

* borderline

5	

						Examples	S					
Properties	13	14	15	16	16 17	18	19	20 21	21	22	23	24
Cells	oben* or	open*	open* open* open* open*	,uado	open*	oben*	* Open	Open				
Recession &	0	0	0	0	0	0	0	0				
FRD (kg/cm ³)							39	34.5 22.8	22.8			
Moulding												:
overall density (kg/m ³)							42.9			44.2	44.2 43.9 44.8	44.8
Core density (kg/m³)	21.1	20.5	,	20.2 19.1 22.3	22.3	22.3	41			42.4	42.4 41.1 41.7	41.7
Compression hardness												,
CLD 40 % (kPa)	1.4	1.6	1.5	1.6	1.5	1.9	3.7	4.6	2.7	4.6 2.7 4.6 5.0		4.6
Hysteresis (%)	3.0	31.9	32.4	34.5		30.4	26.9	40.6	42.9	28.7	\neg	28.4
Compression set (thick)												1
Drv 75 % (%)	12.4	11.6	18.9	25.6	7	7.5	10.1			8.7	9.1	8.5
Humid 75 % (%)	25.6	36.5		53	13.3	12.5	12.8			10.6		9.4
Indentation hardness												
ILD 40 % (N)							211.8			255	264	267
Hysteresis (%)							28.2			30.9	31.7	30.7
Resilience (%)	51.5	51	20	20	26	53.5	55.4			54.6	54.9	54.2
Tear strength												
Max (N/m)	205	219	225	248	175	169	190				249	246
Tensile strength (kPa)	70	81	80	84	78	83	87.2	·		82	79	84
Elongation (%)	1.35	117	123	115	108	105	06				97	95

* borderline

45 ·

			Example	ple		
100000	25	26	27	28	29	30
Component			C	CO	20	٥
Polvol A	90	83	000	2	3	
Polvol H	10	15	20			
Polvol T				10	15	20
Water	4.2	4.2	4.2	4.2	4.2	4.2
R 4113	0.8	0.8	0.8	8.0	0.8	0.8
Niew Al	0.1	0.1	0.1	0.1	0.1	0.1
N 33 LV	0.8	0.8	0.8	0.8	8.0	0.8
Teographs D	09	9	09	09	09	09
NCO index	89	89	89	89	89	89
Cells	oben	uado	oben	oben	oben	oben
* Coissecon	22	16	ഗ	27	20	10
FRD (kg/cm³)	50.5	43.8	36.3	55.9	47.9	37.8
Compression hardness		,	,		•	ים
CLD 40 % (kPa)	7.0	0.9	5.T	7.3	· 0	7.5
Hystoresis (%)	33.9	35.7	39.6	34.1	35.0	37.7

Claims

5		CLAIMS
10	5	1 A polyol composition comprising: bl) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxyl functionality of 2-6 where the EO is present as tipped EO, the EO content being between 10-25 %;
15	10	b2) a polyoxyethylene-polyoxypropylene polyol, having an average nominal hydroxy functionality of 2-6, where the EO is present as tipped EO and random EO, the total EO content being between 20-50 %, the tipped EO
20	15	content being between 10-20 %, b3) a polyol, having an average nominal hydroxy functionality of 2-6, and comprising EO and optionally PO where the EO is present as random EO, the EO content
25	20	being at least 50 %, these polyols b1, b2 and b3 being present according to the following proportions, based on the combined weights of b1, b2 and b3, b1: 60-97 wt %, b2: 3- 40 wt %, b3: 0-25 wt %.
30		2 The polyol composition according to claim 1, which comprises the polyols b1, b2 and b3 according to the following proportions: b1: 65-90 wt %, b2: 10-
35	25	30 wt %, b3 : 0-10 wt %.
40	30	3 The polyol composition according to claim 1 or 2, in which in the polyoxyethylene-polyoxypropylene polyol b2), the weight ratio tipped EO/random EO is between 1:3-3:1.
45		4 The polyol composition of claims 1-3, wherein the polyol b2) is of the -PO-PO/EO-EO type.
	35	5 The polyol composition of claims 1-3, wherei

the polyol b2) is of the -PO/EO-EO type.

50

PCT/EP00/04038 WO 00/73362 21

3		the functionality of the polyols bl, b2 and b3 is 2-4.
		the functionality of the polyoto bi, by and by is I i.
		7 The polyol composition of claims 1-6, wherein
10	5	the polyol b3) is a polyoxyethylene polyol.
		8 The polyol composition of claims 1-7, which
		comprises dispersed particles.
15		
	10	9 The polyol composition of claims 1-8, wherein
		the equivalent weight of polyols b 1) and b 2) is 1000-
		4000 and of polyol b 3) is 200-3000.
20		10 Process for preparing a flexible polyurethane
	15	foam at an NCO index of 70-120 by reacting:
		a) a polyisocyanate composition;
25		bl) a polyoxyethylene-polyoxypropylene polyol,
20		having an average nominal hydroxyl functionality of 2-6
		where the EO is present as tipped EO, the EO content
	20	being between 10-25 %;
30	•	b2) a polyoxyethylene-polyoxypropylene polyol,
		having an average nominal hydroxy functionality of 2-6,
		where the EO is present as tipped EO and random EO, the
	25	total EO content being between 20-50 %, the tipped EC content being between 10-20 %,
35	25	b3) a polyol, having an average nominal hydroxy
		functionality of 2-6, and comprising EO and optionally PO
		where the EO is present as random EO, the EO content
		being at least 50 %,
40	30	these polyols b1, b2 and b3 being present according
		to the following proportions, based on the combined
•		weights of bl, b2 and b3, b1 : 60-97 wt %, b2 : 3-
45		40 wt %, b3 : 0-25 wt %;
70		c) water; and
	35	 d) additives and auxiliaries known per se.

50

55 ,

5		11 The process according to claim 10, in which the polyols b1, b2 and b3 are used according to the following proportions: b1: 65-90 wt %, b2: 10-30 wt %, b3: 0-10 wt %.
10	5	12 The process according to claim 10 or 11, in which in the polyoxyethylene-polyoxypropylene polyol b2), the weight ratio tipped EO/random EO is between 1:3-3:1.
15	10	13 The process of claims 10-12, wherein the polyol b2) is of the -PO-PO/EO-EO type.
20	15	14 The process composition of claims 10-13, wherein the polyol b2) is of the -PO/EO-EO type.
25	15	15 The process of claims 10-14, wherein the functionality of the polyols b1, b2 and b3 is 2-4.
	20	16 The process of claims 10-15, wherein the polyol b3) is a polyoxyethylene polyol.
30		17 The process of claims 10-16, in which the polyol comprises dispersed particles.
35	25	18 The process of claims 10-17 wherein polyols b 1) and b 2) have an equivalent weight of 1000-4000 and polyol b 3) of 200-3000 and the polyisocyanate is diphenylmethane diisocyanate optionally comprising
40 ·	30	homologues thereof having an isocyanate functionality of 3 or more and modified variants thereof.
4 5		19 A reaction system comprising A) a polyisocyanate and B) an isocyanate-reactive component comprising the polyol composition of claims 1-8 and
	35	water.